

Universal Néel Temperature in Three-Dimensional Quantum Antiferromagnets

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We study three-dimensional dimerized $S = 1/2$ Heisenberg antiferromagnets, using quantum Monte Carlo simulations of systems with three different dimerization patterns. We propose a way to relate the Néel temperature T_N to the staggered moment m_s of the ground state. Mean-field arguments suggest $T_N \propto m_s$ close to a quantum-critical point. We find an almost perfect universality (including the prefactor) if T_N is normalized by a proper lattice-scale energy. We show that the temperature T^* at which the magnetic susceptibility has a maximum is a good choice, i.e., T_N/T^* versus m_s is a universal function (also beyond the linear regime). These results are useful for analyzing experiments on systems where the spin couplings are not known precisely, e.g., TiCuCl_3 .

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Quantum fluctuation can drive continuous phase transitions between different kinds of ground states of many-body systems. While transitions taking place at temperature $T > 0$ are controlled by thermal fluctuations, quantum fluctuations also play a role here. Quantum-critical scaling can often be observed throughout a wide region (the quantum-critical “fan”) extending out from the quantum-critical point ($g_c, T = 0$) into the plane ($g, T > 0$) [1–4], where g is the parameter tuning the strength of the quantum fluctuations. In addition, the quantum fluctuations of course also strongly affect the critical temperature T_c , because $T_c \rightarrow 0$ as $g \rightarrow g_c$. One can regard the quantum fluctuations as reducing the order at low temperature ($T \ll T_c$), with the thermal fluctuations eventually destroying it as $T \rightarrow T_c$, but precisely how the two kinds of fluctuations act in conjunction with each other to govern T_c is not known in general.

We will here discuss manifestations of the interplay of quantum and thermal fluctuations for $0 < T < T_N$ in three-dimensional (3D) $S = 1/2$ quantum antiferromagnets with Heisenberg interactions. In these systems one can vary the critical Néel-ordering temperature, T_N , and ultimately achieve a quantum phase transition ($T_N \rightarrow 0$), by considering dimerized couplings, such that each spin belongs exactly to one dimer and the intra- and inter-dimer couplings are different. The Hamiltonian for such models can be generically written as

$$H = J_1 \sum_{\langle i,j \rangle_1} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle i,j \rangle_2} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

where $\langle i,j \rangle_a$ denotes a pair of spins coupled at strength J_a , with $a = 1$ and $a = 2$ corresponding to inter- and intra-dimer bonds, respectively. Three examples of such dimerized 3D lattices are shown in Fig. 1. In (a) and (b) the spins form simple cubic lattices, and each nearest-neighbor site pair is coupled either by J_1 or J_2 . In (c) two different cubes each have all J_1 couplings, and pairs of spins in different cubes form the J_2 -coupled dimers. We will use the ratio $g = J_2/J_1$ as the tuning parameter. When $g \approx 1$ the system is Néel-ordered at $T = 0$

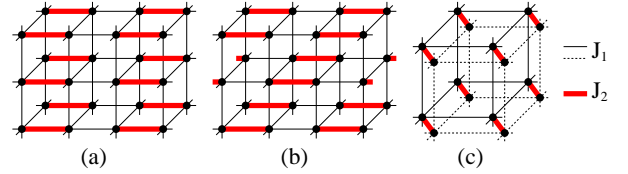


FIG. 1: (Color online) Dimerized 3D lattices; (a) columnar dimers, (b) staggered dimers, and (c) double cube. For a system of length L , the number of spins is $N = L^3$ in (a) and (b), and $N = 2L^3$ in (c). The two different coupling strengths J_1 and J_2 are indicated by thin (black dashed and solid) and thick (red) lines, respectively.

and when $g \rightarrow \infty$ it decouples to form a set of independent dimers, with the ground state becoming a trivial quantum paramagnet with a singlet-product ground state. The system for any $J_1 > 0$ and $J_2 > 0$ is accessible to unbiased numerical studies with efficient quantum Monte Carlo (QMC) methods with loop updates [5–7].

Analogous dimerized Heisenberg models have been studied extensively with QMC in two dimensions, where there is order only at $T = 0$ (for $g < g_c$) and the nature of the quantum-critical point and its associated scaling fan has been the main focus of interest [7, 8]. Some simulations have also been previously carried out for 3D dimerized models [9–11]. Here we report calculations uncovering universal aspects of the ordering temperature, from systems close to the quantum-critical point to deep inside the Néel phase. We develop a scaling procedure of direct relevance to experiments. Our results also provide new insights into the relevant energy scales present in the 3D Néel state and demonstrate an effective decoupling of thermal and quantum fluctuations.

Experimental issues.—The best experimental realization so far of a dimerized system with a quantum phase transition is TiCuCl_3 under pressure [12–14]. The spin dimers here form on pairs of Cu atoms that can clearly be identified as the most strongly coupled neighbors. The inter-dimer couplings are, however, more complicated than in the simple nearest-neighbor Hamiltonian (1). There are several significant exchange constants but

their exact values are not known (although they have been estimated based on approximate calculations of the magnon dispersion, which can be compared with experiments [12, 15]). The dimers nevertheless form a 3D network, and one can expect the same ground state phases and phase transitions as with the simplified Hamiltonian (1). Under ambient pressure, TiCuCl_3 exhibits no magnetic order, but beyond a critical pressure antiferromagnetic order emerges continuously. The interpretation of this is that one or several of the inter-dimer couplings become strong enough for Néel order to form. The observed longitudinal and transversal excitation energies agree well with predictions based on $O(3)$ symmetry breaking and Goldstone modes [15, 16].

The fact that the microscopic spin-spin couplings in TiCuCl_3 , and how they depend on pressure, are not known accurately is a complication when comparing experimental results with calculations for a specific model Hamiltonian. In this situation it is useful to make comparisons that do not require any explicit knowledge of the couplings. Here we will investigate how the Néel temperature is related to the staggered magnetization m_s at $T = 0$. Based on unbiased QMC calculations for the three different dimerized models defined in Eq. (1) and Fig. 1, we show that the curve $T_N(m_s)$ exhibits a remarkable universality when properly normalized, not just close to the quantum-critical point but extending to strongly ordered systems. Our results give a parameter-free scaling function that can be compared with experiments.

Quantum Monte Carlo calculations.—We have used the stochastic series expansion (SSE) QMC method with very efficient loop updates [5–7] to calculate the squares $\langle m_z^2 \rangle$ and $\langle m_{sz}^2 \rangle$ of the z -components of the uniform and staggered magnetizations,

$$m_z = \frac{1}{N} \sum_{i=1}^N S_i^z, \quad m_{sz} = \frac{1}{N} \sum_{i=1}^N \phi_i S_i^z, \quad (2)$$

where the phases $\phi_i = \pm 1$ correspond to the sublattices of the bipartite systems in Fig. 1. The uniform susceptibility is $\chi = \langle m_z^2 \rangle / (TN)$. We also study the Binder ratio, $R_2 = \langle m_{sz}^4 \rangle / \langle m_{sz}^2 \rangle^2$, and the spin stiffness constants ρ_s^α in all lattice directions ($\alpha = x, y, z$), $\rho_s^\alpha = d^2 E(\theta_\alpha) / d\theta_\alpha^2$, where E is the internal energy per spin and θ_α a uniform twist angle imposed between spins in planes perpendicular to the α axis. The stiffness constants can be related to winding number fluctuations in the simulations [7].

We use standard finite-size scaling [7] to extract T_N . At T_N , the stiffness constants scale with the system length as $\rho_s^\alpha \propto L^{2-d}$, where the dimensionality $d = 3$. Thus, $\rho_s^\alpha L$ should be size-independent at T_N , while this quantity vanishes (diverges) for $T > T_N$ ($T < T_N$). In practice, this means that curves versus T (at fixed g) for two different system sizes L cross each other at a point which drifts (due to scaling corrections) toward T_N with increasing L . The dimensionless Binder ratio also has

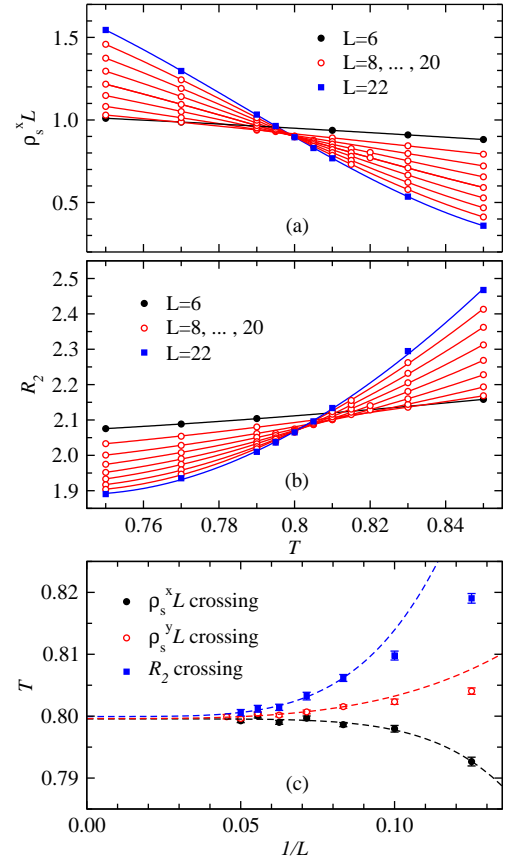


FIG. 2: (Color online) Procedures used to extract the critical temperature T_N . (a) and (b) show $\rho_s^x L$ and R_2 , respectively, for the columnar dimer model at coupling ratio $g = 3.444$. The error bars are smaller than the symbols. Using polynomial fits to data for two lattice sizes, L and $L + 2$, crossing points between the curves are extracted. Results are shown in (c), along with fits of the form $T_N(L) = T_N(\infty) + a/L^w$ (to the large- L data for which this form obtains). Extrapolations of the three quantities give $T_N = 0.7996(3)$, $0.7996(6)$, and $0.7999(5)$ for $L \rightarrow \infty$, all consistent with each other within errors bars.

this kind of behavior and provides us with a different T_N estimate to check for consistency. Figs. 2(a,b) show examples of these crossing behaviors for $\rho_s^x L$ and R_2 . The crossing points drift in different directions and bracket T_N . Fig. 2(c) shows the L dependence of crossing points extracted from data for $(L, L+2)$ system pairs, for R_2 and two different stiffness constants. Power-law fits are used to extrapolate to infinite size. The mutual consistency of the T_N value so obtained using different quantities gives us confidence in the accuracy of this procedure.

To extract the $T = 0$ sublattice magnetization, we carry out simulations at temperature $T = J_1/L$. Note that, in a Néel phase with $T_N > 0$, any $T(L)$ such that $T(L \rightarrow \infty) \rightarrow 0$ can be used for extrapolations to the thermodynamic limit and $T = 0$. Our choice is a natural way to scale the temperature since the lowest spin waves have energy $\propto 1/L$. We also did some calculations

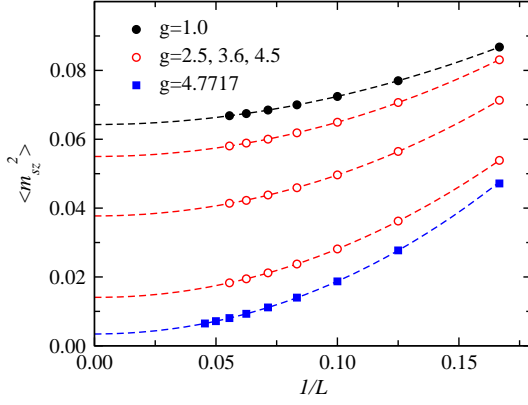


FIG. 3: (Color online) Extrapolation of the sublattice magnetization obtained in simulations with $T = J_1/L$ of the double-cube Heisenberg model at different coupling ratios g . The error bars are much smaller than the symbols. The fitting function used for $L \rightarrow \infty$ extrapolations is $a + b/L^2 + c/L^3$ (where we exclude the linear term because it comes out very close to zero in fits including it).

with $T = 1/2L$ and obtained consistent extrapolated results. Examples of the L dependence are shown in Fig. 3 for the double-cube model at several different coupling ratios. Taking into account rotational averaging in spin space, the final result for the sublattice magnetization is given by the $L \rightarrow \infty$ extrapolated $\langle m_{sz}^2 \rangle$ (for which we use a polynomial fit, as shown in Fig. 3); $m_s = \sqrt{3\langle m_{sz}^2 \rangle}$.

Universality of T_N versus m_s .—Following the above procedures, we have calculated T_N and m_s accurately for all three dimer models at several coupling ratios g , from close to g_c to deep inside the Néel phase. We graph T_N versus m_s in Fig. 4. T_N is scaled by three different energy units; the inter-dimer coupling J_1 in (a), the sum of couplings J_s connected to each spin in (b), and the temperature T^* at which the susceptibility exhibits a peak in (c). Before discussing these normalizations of T_N in detail, let us examine the reason for the linear behavior, $T_N \propto m_s$, seen in the QMC results for small [and in (b),(c) even quite large] m_s .

A semi-classical mean-field argument (inspired by the “renormalized classical” picture developed in two dimensions [1]) leading to $T_N \propto m_s$ is the following: To compute T_N in a classical system of spins of length S , one replaces the coupling of a spin \mathbf{S}_0 to the total spin of its neighbors δ , $J \sum_{\delta} \mathbf{S}_{\delta}$, by the thermal average $J \sum_{\delta} \langle \mathbf{S}_{\delta} \rangle$. In the presence of quantum fluctuations, this mean field seen by \mathbf{S}_0 is reduced, which is taken into account by a renormalization; $\langle \mathbf{S}_{\delta} \rangle \rightarrow (m_s/S) \langle \mathbf{S}_{\delta} \rangle$. The thermal fluctuations are, thus, added on top of the quantum fluctuations at $T = 0$, under the assumption that the quantum effects will not change appreciably for $T > 0$ (i.e., the thermal fluctuations are assumed to be solely responsible for further reducing the order). Note that \mathbf{S}_0 should not be renormalized here, but is computed as a thermal expectation value and should satisfy the self-consistency

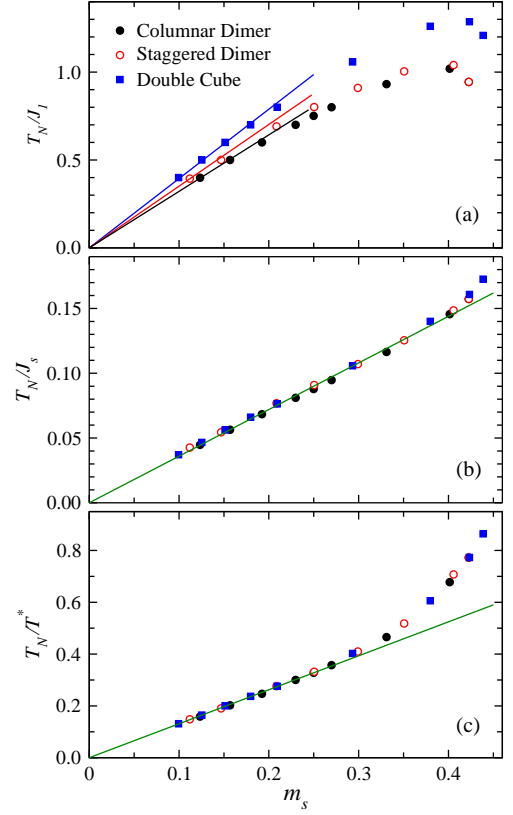


FIG. 4: (Color online) The Néel temperature T_N versus the sublattice magnetization for the three different dimerized models and with T_N normalized in three different ways. T_N is measured in units of (a) the inter-dimer coupling J_1 , (b) the total coupling J_s per spin, (c) the peak temperature T^* of the susceptibility. A linear dependence obtains in all cases for small to moderate m_s , as indicated by fitted lines. Note that $m_s \leq 1/2$ for $S = 1/2$.

condition $\langle \mathbf{S}_{\delta} \rangle = \langle \mathbf{S}_0 \rangle$. The final magnetization curve is given by $(m_s/S) \langle \mathbf{S}_0 \rangle$. In this procedure of decoupling the classical and quantum fluctuations, one clearly effectively has $J \rightarrow (m_s/S)J$ and, thus, $T_N \propto m_s$.

The assumption that the quantum renormalization factor m_s/S is T -independent up to T_N can be valid only if T_N is small. The energy scale in which to measure T_N when stating this condition should be dictated by the spin-wave velocity, which stays non-zero at the quantum-critical point [17] [i.e., not by the long-distance energy scale $\rho_s(T = 0)$, which vanishes as $g \rightarrow g_c$ and is unrelated to the density of thermally excited spin waves]. A linear dependence is seen in Fig. 4 up to rather large values of m_s (where $T_N \sim J_1$). A linear dependence was also recently found in the columnar dimer model based on high- T expansions [18] (with much larger error bars).

Returning now to the issue of how to best normalize T_N , we note that in Fig. 4(a), where the inter-dimer coupling J_1 is used, the curve for the double-cube model is significantly above the other two. This is clearly because the constant J_1 does not account for the different aver-

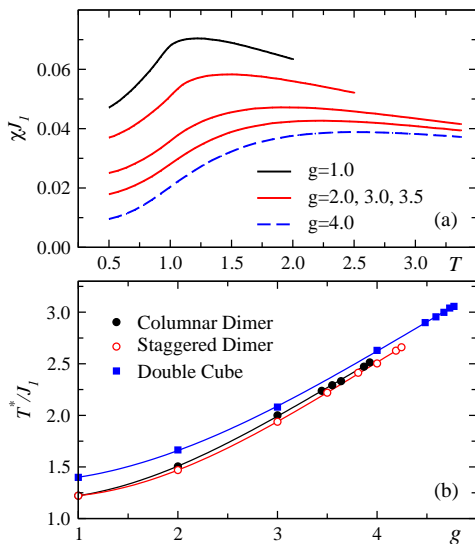


FIG. 5: (Color online) (a) Susceptibility versus temperature of the staggered dimer model at different coupling ratios. The system size is $L = 12$, for which the peak height and location are already $L \rightarrow \infty$ converged. (b) The peak temperature versus the coupling ratio for the three different models.

age couplings in the models. Using instead the sum J_s of couplings connected to each spin, i.e., $J_s = 5 + g$ for the columnar and staggered dimers and $6 + g$ for the double cube (setting $J_1 = 1$), the curves, shown in Fig. 4(b), collapse almost on top of each other. Note that also the curves for the columnar and staggered dimers are closer to each other than in Fig. 4(a), although they have the same definition of J_s . This can be the case because J_s rescales the curves non-uniformly, since $m_s(g)$ and, therefore, $J_s(m_s)$, is different for the two models. The linearity of T_N/J_s versus m_s is also much clearer than before and extends all the way up to $m_s \approx 0.3$.

Although the data collapse is already quite good in T_N/J_s , we can do even better when normalizing with a physical quantity that measures the effective lattice-scale energy. One such energy scale in antiferromagnets is the temperature at which the uniform magnetic susceptibility χ exhibits a peak. This peak is due to the cross-over from the high- T Curie form to the low- T weakly temperature dependent form typical of antiferromagnets. The peak temperature T^* , thus, reflects the short-distance energy scale at which antiferromagnetic correlations become significant. T^* is often used experimentally to extract the value of the exchange constant, using, e.g., the “Bonner-Fisher” curve in one dimension [19]. In spatially anisotropic systems such as the dimerized models we consider here, a natural assumption is that T^* reflects an effective average coupling. In Fig. 5(a) we show examples of the susceptibility close to its peak, and in (b) we show the dependence of T^* on g for all three models. Normalizing T_N with T^* leads to remarkably good data collapse, as shown in Fig. 4(c). Deviations from a common curve are barely detectable. Although we can-

not prove that this function is really universal for all 3D networks of dimers, the results are very suggestive of this.

Discussion.—The universal behavior implies that the $T > 0$ disordering mechanism in the 3D Néel state is completely governed by a single lattice-scale energy (which, as we have shown here, can be taken as the peak temperature T^* of the susceptibility) and the $T = 0$ sublattice magnetization m_s . The extended linear behavior seen in Figs. 4(b,c) shows that the quantum and classical fluctuations at $T < T_N$ are completely decoupled all the way from $g = g_c$ (excluding g_c itself, where $T_N = 0$) to quite far away from the quantum-critical point. Depending on a lattice-scale energy instead of the quantum-critical spin stiffness, the linear behavior is not fundamentally a quantum-critical effect. We have discussed the linearity and decoupling of the fluctuations in terms of a semi-classical mean-field theory, the validity of which implies that the quantum-critical regime [2] commences only above T_N . Deviations from linearity at larger m_s show that the quantum fluctuations are affected (become T -dependent) here, due to the high density of excited spin waves as $T \rightarrow T_N$ because T_N is high. It is remarkable that this coupling of quantum and classical fluctuations also takes place in an, apparently, universal fashion for different systems. It would be interesting to explain this more quantitatively, by deriving the full function T_N versus m_s analytically. Progress in the linear regime has been made recently in work parallel to ours [20].

From a practical point of view, the data collapse of T_N/T^* versus m_s is very useful, because all the quantities involved can be measured experimentally and do not rely on microscopic details. The universal curve can be used to test the 3D Heisenberg scenario without adjustable parameters. The universality likely applies not only to dimer networks, but also to systems where the quantum fluctuations are regulated in other ways.

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